

Shape transformations in rotating ferrofluid drops

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Floating drops of magnetic fluid can be brought into rotation by applying a rotating magnetic field. We report theoretical and experimental results on the transition from a spheroid equilibrium shape to non-axisymmetrical three-axes ellipsoids at certain values of the external field strength. The transitions are continuous for small values of the magnetic susceptibility and show hysteresis for larger ones. In the non-axisymmetric shape the rotational motion of the drop consists of a vortical flow inside the drop combined with a slow rotation of the shape. Nonlinear magnetization laws are crucial to obtain quantitative agreement between theory and experiment.

PACS numbers: 47.20.Hw, 47.55.Dz, 75.50.Mm

The equilibrium shapes of rotating fluid bodies are of importance in various fields of physics as, e.g., astrophysics [1], nuclear fission [2], plasma [3], and biological physics [4]. The famous controversy between Newton and Cassini on whether the Earth has the form of an oblate or prolate rotational ellipsoid started a series of ingenious investigations of the equilibrium shapes of heavenly bodies including work by Maupertius, MacLaurin, Jacobi, Riemann, Poincaré and others. Among the surprising results discovered are the possibility of three-axes ellipsoids as shown by Jacobi and the emergence of pear-shaped configurations found by Poincaré. In rotating non-neutral plasmas, laser cooled in a Penning trap, a novel equilibrium state with non-axisymmetric surface has recently been observed [5]. Tank-treading elliptical membranes in a shear flow have been used as a theoretical model for the motion of human red blood cells [6].

Experimental investigations of the stationary shapes of rotating bodies in the laboratory usually start with a static drop of fluid floating in another, immiscible fluid of the same density. The rotational motion of the drop is then set up by, e.g., using a rotating shaft [7] or applying an acoustic torque [8]. An elegant way to spin up drops made from polarizable fluids is to use rotating electric [5] or magnetic fields [9, 10].

In the present letter we report theoretical and experimental investigations of rotating ferrofluid drops and in particular perform the first quantitative study of a transition from a spheroidal to a non-axisymmetric equilibrium shape in this system. Moreover, providing an approximate solution of the hydrodynamic flow problem inside and outside the drop we are able to analyze the rotational motion of the drop shape and to separate it from the internal hydrodynamic flow.

Ferrofluids are suspensions of ferromagnetic nanoparticles in suitable carrier liquids combining the hydrodynamic behaviour of a Newtonian fluid with the magnetic properties of a super-paramagnet [11]. A rotating external field induces a rotational motion of the nano-

particles which due to their viscous coupling to the surrounding liquid transfer the angular momentum to the whole drop. The system has been studied previously by Bacri et al. using microdrops with a typical radius of 10 μm , very small surface tension and a viscosity much larger than that of the outer fluid [9, 12]. Although an instability of the axisymmetric shape was found experimentally and theoretically [13] the emerging non-axisymmetric configurations could not be studied quantitatively since for the given parameter values very irregular shapes arise. In our experiments we used much larger drops of radius $R = 2.75\text{mm}$ of a kerosine-based ferrofluid with magnetic susceptibility $\mu_r = 15.3$ and dynamic viscosity $\eta_1 = 0.019\text{ Pas}$ immersed in 3-bromo-1,2-propandiol with dynamic viscosity $\eta_2 = 0.058\text{ Pas}$. The interface tension is $\sigma = 2.8 \cdot 10^{-3}\text{ N/m}$, the frequency of the magnetic field 560 Hz. For these parameter combinations we find well-defined transitions to three-axes ellipsoids which can be analyzed in detail.

From the parameter values given above we find for the typical time of shape relaxations of the drop $\tau_s \cong 0.1\text{s}$. We are thus considering the case of a fast rotating field, $\omega\tau_s \gg 1$, in which the drop assumes an oblate shape with the short axis perpendicular to the field plane. Its form is very near to an ellipsoid and we will use this approximation throughout our theoretical analysis. The shape will be specified by the ratios $\epsilon_b = a/b$ and $\epsilon_c = a/c$ between the seminaxes of the ellipsoid where as usual $a \geq b \geq c$ is assumed.

Estimating the orders of magnitude of the energies due to magnetization, surface tension, viscosity and inertia, respectively, we find that for the experimentally relevant parameter values the shape is almost completely determined by the balance between surface and magnetic energy where the latter is conveniently averaged over one period of the field rotation. The calculation of the magnetic energy is simplified by the fact that an ellipsoid in a homogenous external field builds up a *homogenous* magnetization. Using a coordinate system in which the

external field is of the form $\mathbf{G} = (G \cos(\omega t), G \sin(\omega t), 0)$ this magnetization is determined by the relations

$$G_x = H_x + n_x M_x, \quad G_y = H_y + n_y M_y \quad (1)$$

between the components of the external field \mathbf{G} , the internal field \mathbf{H} and the magnetization \mathbf{M} [14]. Here n_x and n_y denote the demagnetization factors along the x and y axis, respectively, which are known functions of ϵ_b and ϵ_c [14].

The calculation of the magnetic energy is most easily accomplished assuming a linear magnetization law, $\mathbf{M} = \chi \mathbf{H}$, with the susceptibility $\chi = \mu_r - 1$. We then find for the magnetic energy $E_m = -\mu_0 V \mathbf{M} \cdot \mathbf{G}/2$ using (1)

$$E_m(t) = -\frac{\mu_0 V}{2} \chi G^2 \left(\frac{\cos^2(\omega t)}{1 + \chi n_x} + \frac{\sin^2(\omega t)}{1 + \chi n_y} \right) \quad (2)$$

with V denoting the volume of the drop. Averaging over one period of the rotation [15], using the well-known expression for the surface of a three-axes ellipsoid, and observing volume conservation for the drop we obtain the following result for the sum E of magnetic and surface energies

$$\frac{E}{2\pi\sigma R^2} = -\frac{\chi}{6} B \left(\frac{1}{1 + \chi n_x} + \frac{1}{1 + \chi n_y} \right) + \epsilon_b^{2/3} \epsilon_c^{-4/3} \left[1 + \frac{\epsilon_c}{\epsilon_b \sqrt{\epsilon_c^2 - 1}} (F(m, \kappa) + (\epsilon_c^2 - 1)E(m, \kappa)) \right]$$

Here F and E are elliptic integrals of first and second kind, respectively, [16], $m = \sqrt{\epsilon_c^2 - 1}/\epsilon_c$, $\kappa = \sqrt{(\epsilon_c^2 - \epsilon_b^2)/(\epsilon_c^2 - 1)}$, and $B = \mu_0 G^2 R/\sigma$ is the magnetic Bond number measuring the strength of the external field. Minimizing this expression numerically in the geometry parameters ϵ_b and ϵ_c the dependence of the shape of the drop can be determined for varying external field strength G . Results for the parameter values given above are shown in figs.1 and 2 together with our experimental findings.

The main result is a transition from a spheroid characterized by $\epsilon_b = 1$ to a pronounced non-axisymmetric form of a *three-axes ellipsoid* for intermediate values of the magnetic field strength. This transition takes place only if the magnetic permeability is large enough, $\mu_r \gtrsim 5.1$. It occurs via supercritical bifurcations up to $\mu_r \cong 11.6$ and through subcritical transitions with hysteresis for still higher values of μ_r including our experimental value.

From the figures it is seen that the linear theory is in qualitative agreement with the experiment. It also yields quantitatively good results for the values of the Bond number at which the transitions to three-axes ellipsoids occur. The ratios between the semiaxes, however, are overestimated, with the discrepancy increasing with the field strength. In fact the magnetic field corresponding to

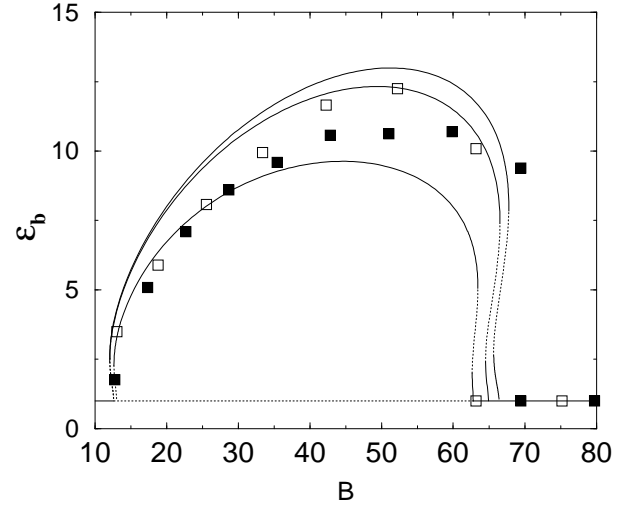


FIG. 1: Ratio $\epsilon_b = a/b$ between the two largest semiaxes of a rotating ferrofluid drop as a function of the magnetic Bond number B . Squares are experimental values with filled symbols corresponding to increasing, empty symbols to decreasing field strength, respectively. The curves show from top to bottom the results for a linear magnetization law, $\mathbf{M} = \chi \mathbf{H}$, for the Langevin $M(H)$ and for the dynamic curve $M(H)$ as determined from an independent experiment. Full lines correspond to stable configurations, dotted lines to unstable ones. There are no fit parameters.

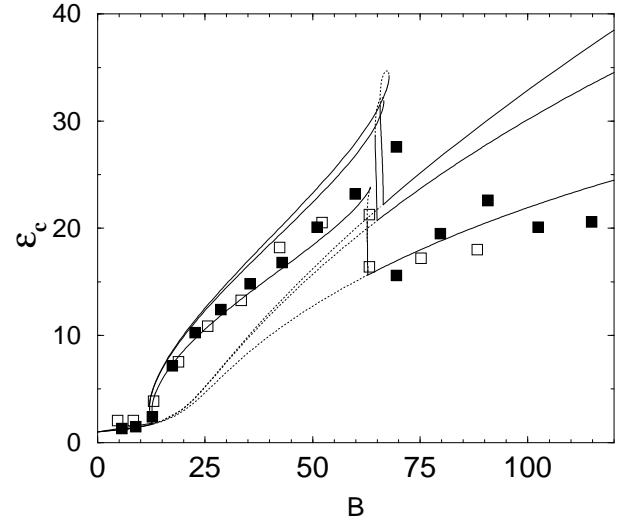


FIG. 2: Same as fig.1 for the ratio $\epsilon_c = a/c$ between the largest and the smallest semiaxes of the rotating drop.

a Bond number of $B \cong 80$ is of the order of the saturation magnetization $M_\infty \cong 80 \text{ kA/m}$ and hence deviations from the linear law $\mathbf{M} = \chi \mathbf{H}$ become important.

In order to account for these deviations from a linearity we have extended the theory to general magnetization curves $M(H)$. The field inside the ellipsoid is still homo-

geneous and using (1) we get

$$\frac{1}{G^2} = \frac{\cos^2(\omega t)}{(H + n_x M(H))^2} + \frac{\sin^2(\omega t)}{(H + n_y M(H))^2} \quad (3)$$

from which for given $M(H)$ the time dependence of the internal field H can be determined numerically. The magnetic energy can now be written in the form

$$E_m(t) = -\frac{\mu_0 V}{2} \left[G^2 M \left(\frac{\cos^2(\omega t)}{H + n_x M} + \frac{\sin^2(\omega t)}{H + n_y M} \right) - MH + 2 \int_0^H dH' M(H') \right]. \quad (4)$$

For a linear magnetization law the last two terms cancel and we find back (2). Expression (4) is numerically averaged over one period of the field rotation and the total energy is minimized with respect to ϵ_b and ϵ_c .

A standard choice for the magnetization curve of a ferrofluid is the Langevin law $M = M_\infty(\coth(\xi) - 1/\xi)$ with $\xi = 3\chi H/M_\infty$. The corresponding results are included in figs.1 and 2. It is clearly seen that although the values for ϵ_b and ϵ_c are reduced in comparison with the linear theory the corrections are rather small. The physical reason for the remaining discrepancy lies in relaxation effects of the magnetization which for the comparatively high frequency of the external field start to play a role. We have therefore determined the magnetization curve $M(H)$ of our ferrofluid for an oscillating magnetic field from an independent experiment. The lowest curves in figs.1 and 2 result from the use of a spline interpolation of these data in our numerical code. Whereas the experimental results for ϵ_b are now underestimated for large values of B the correspondence between theory and experiment for ϵ_c is rather satisfactory.

We now turn to the analysis of the drop motion. In the experiments we observe a slow rotation of the non-axisymmetric drop shape with angular velocity $\Omega \cong 0.1\text{Hz}$ in the direction of the field rotation. In order to discuss this motion theoretically we have to solve the hydrodynamic flow problem inside and outside the drop taking into account the appropriate boundary conditions for normal and tangential stresses at the surface. In its general form this is a formidable free boundary value problem. We hence employ some reasonable approximations consistent with our assumption of an elliptical drop shape. For the experimentally relevant sizes and viscosities the inertial terms in the hydrodynamic equations are small and we may use the Stokes approximation for the determination of the flow fields. It is convenient to use the coordinate system in which the shape of the drop is at rest. In this frame we assume that the motion inside the drop is of uniform vorticity ζ and hence use for the internal and external flow fields the ansatzes

$$\mathbf{v}^{\text{in}} = (-\zeta \frac{ya}{b}, \zeta \frac{xb}{a}, 0), \quad \mathbf{v}^{\text{ex}} = \mathbf{v}^{\text{in}} + (u_J, v_J, w_J). \quad (5)$$

From the continuity of the velocities at the surface of the drop we infer that (u_J, v_J, w_J) describes the flow field outside a *rigid* elliptical particle at rest in a viscous fluid with asymptotic velocity $\lim_{r \rightarrow \infty} (u_J, v_J, w_J) = ((\Omega + \zeta a/b)y, -(\Omega + \zeta b/a)x, 0)$, a problem solved by Jeffery many years ago [17].

To complete the solution in the discussed approximation we need two equations to fix the so far undetermined parameters Ω and ζ . As first equation we use the balance between the viscous torque experienced by the drop in its rotational motion and the magnetic torque, again averaged over one period of the driving. The second one results from the stationarity of the energy balance stating that the average work done by the external field per unit time must be equal to the energy dissipated per unit time in the viscous flows. This latter requirement can be rewritten as a continuity condition for the tangential stress *averaged* over the surface of the drop [18, 19].

The expression for the viscous torque builds on Jeffery's solution and can be obtained in the same way as for a slowly rotating field, $\omega\tau_s \ll 1$, [20]. For the determination of the magnetic torque $\mathbf{L} = \mu_0 V (\mathbf{M} \times \mathbf{G})$ it is crucial to take into account the finite relaxation time of the magnetization since a non-zero averaged torque results from the phase lag between \mathbf{M} and \mathbf{H} . The explicit calculation is again feasible analytically for a linear magnetization curve $\mathbf{M} = \chi \mathbf{H}$ where the susceptibility is now complex, $\chi = \chi_1 - i\chi_2$. We will assume $\chi_2 \ll \chi_1$ consistent with the experiment where $\chi_1 \cong 14.3$ and $\chi_2 \cong 2.1$. Using (1) and $\omega - \Omega \cong \omega$ we get for the z -component of the averaged magnetic torque

$$\overline{L_z} = \frac{\mu_0 V G^2}{2} \chi_2 \left(\frac{1}{(1 + \chi n_x)^2} + \frac{1}{(1 + \chi n_y)^2} \right). \quad (6)$$

In the integrated balance of tangential stresses the viscous contributions can be obtained from Jeffery's solution [17, 18]. The tangential part of the Maxwell stress tensor of the magnetic field is automatically continuous at the interface due to the boundary conditions obeyed by the fields [14]. However, in the present non-equilibrium situation there *is* a magnetic contribution due to the additional term $(M_i H_k - M_k H_i)/2$ in the stress tensor introduced by Shliomis [21]. Taking into account the various contributions two equations for Ω and ζ can be derived. The final expressions are rather long and will be published elsewhere [22].

Results for the rotation frequency Ω of the drop are shown in fig.3 together with the corresponding experimental findings. The linear theory overestimates the magnetic torque and therefore also the frequency of rotation. To investigate the effects of deviations from the linear magnetization curve we have calculated the magnetic torque using the numerical solution of the magnetization relaxation equation [23]

$$\frac{d\mathbf{M}}{dt} = -\frac{1}{\tau_{\parallel} H^2} (\mathbf{H} \cdot (\mathbf{M} - \mathbf{M}_0)) \mathbf{H} - \frac{1}{\tau_{\perp} H^2} \mathbf{H} \times (\mathbf{M} \times \mathbf{H}).$$

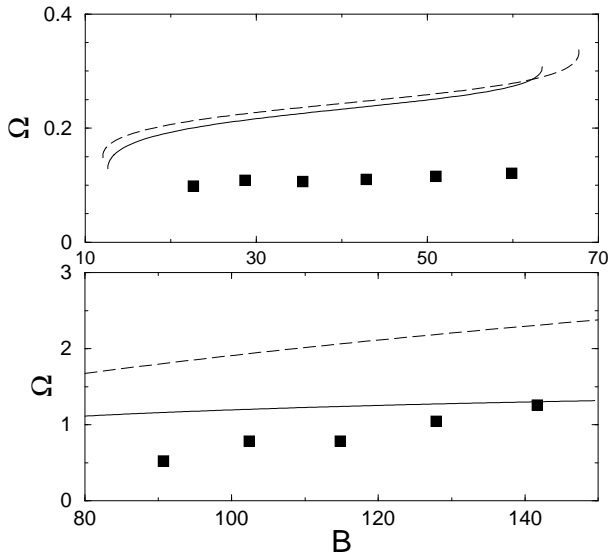


FIG. 3: Rotation frequency of the drop as function of the magnetic Bond number. Squares are experimental values, lines are theoretical results using a linear magnetization law (dashed) and the magnetization curve $M(H)$ as determined in an independent experiment (full), respectively. The upper part describes the rotation of the three-axes ellipsoid, the lower one the hard body rotation of a disk.

The static magnetization $\mathbf{M}_0(H)$ and the field dependence of the relaxation times τ_{\parallel} and τ_{\perp} are determined from independent experiments, the geometry of the drop is taken from figs.1 and 2. Although the magnetic torque is substantially smaller than in the linear case the results for the rotation period of the non-axisymmetric drop are almost the same. This is due to the reduced viscous torque resulting from smaller values of ϵ_b and ϵ_c in the nonlinear theory. The remaining differences with the experiment may be due to the polydispersity of the fluid requiring a whole spectrum of relaxation times and a small ellipticity of the external field. These questions will be dealt with in detail elsewhere [22].

For the axisymmetric state, $\epsilon_b = 1$, shown in the lower part of fig.3 the situation is somewhat simpler. In this case the drop forms a flat disk rotating like a hard body. For large values of B this disk develops peaks around its perimeter due to the normal field instability [9, 11, 13] making an experimental determination of the rotation frequency easy. As can be seen from fig.3 the linear theory again strongly overestimates the rotation frequency whereas the non-linear theory yields substantially smaller results. For Bond numbers between 80 and 100 the disk shows few, rather large peaks resulting in an increased viscous torque and therefore a slower rotation.

For larger values of the Bond number the peaks get smaller and more numerous and the approximation of the shape by an ellipsoid becomes again more accurate resulting in a reasonable agreement between theory and experiment.

We would like to thank Marcus Hauser for his help in procuring some of the chemicals needed for the experiments. Financial support from the *Deutsche Forschungsgemeinschaft* under grant FOR 301/2-1 is gratefully acknowledged.

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